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TITLE OF PROJECT

AASERT STUDENT RESEARCH ON NON-OXIDE STRUCTURAL CERAMICS-ALLOY DESIGN FOR IMPROVED SINTERABILITY AND MECHANCAL PERFORMANCE

FiINAL REPORT

December 31, 1998

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PRINCIPAL INVESTIGATOR

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II. *OBJECTIVES*

The objective of this research is to design microstructures in non-oxide ceramics to improve their mechanical performance and sinterability. Silicon nitride and silicon carbide with similarly controlled microstructures and residual stress distributions ~~will~~ have ~~be~~ been investigated.

III. *STATUS OF EFFORT*

Abstract

The grant was initiated in 1996. The AASERT student supported by the grant, Anatoly Rosenflanz, discovered an in-situ toughened α'-SiAlON and a patent was filed. This research has been publicly reported since 1997 and has received broad attention. We have also explored the compositional range over which in-situ toughened α'-SiAlON can be obtained. This essentially encompasses the entire single α'-phase region at the higher temperature. We have made a systematic effort to obtain kinetic data on phase transformation at different compositions, since this knowledge is critically important for the precise control of nucleation and growth which dictate the microstructure. We have shown that the mechanical strength of α'-SiAlON can exceed 1 GPa and the strength retention is good at least up to 1350°C. We have lastly reported phase relations in melilite-containing silicon nitride and their properties, as well as a series of superplastic silicon nitride. ~~This grant is now closed.~~

IV. *ACCOMPLISHMENTS/NEW FINDINGS*

<u>Nucleation Control of Microstructure</u>

The essence of microstructure design that enables the formation of in-situ toughened a'-SiAlON is to control nucleation of α' phase so that relatively few nuclei compete for growth. There are three general ways to achieve this goal. First, starting powders can be chosen to be energetically more stable or crystallographically less similar to the product phase. This implies that β- Si_3N_4 powders are better as the starting powders. The second method is to choose a composition with less stability for the α' phase. This dictates the choice of larger cations or compositions near the phase boundary. An extension of the second approach is to take advantage the temperature dependence of the phase stability. This dictates the use of lower temperature for nucleation. We have demonstrated that these three approaches, individually or in combination, with various conceivable variations, can be practiced to render any single phase α'-SiAlON composition amenable to obtaining a fibrous microstructure.

Detailed kinetic studies have been concentrated in different compositional regions since the rates of nucleation and phase transformation are strongly dependent on the driving force and the viscosity, and they must be determined for different cations and different O/N ratios. So far, we have obtained complete data in the Si_3N_4 rich corner, and are in the process of obtaining similar data in the AlN rich corner. Since the aim is to control nucleation, such data were obtained under experimental conditions when only very fine grains form. The results obtained thus far can be interpreted using a

simple picture, namely the kinetics are directly correlated to the magnitude of the driving force. In addition, the effect of starting powders and reaction pathway in general has been studied and examined using the concept of driving force. We are able to use this picture to rationalize all the microstructure observations following the argument of nucleation and growth. The success of this simple picture is important since the α'-SiAlON systems are chemically complicated. Without such a simple guiding principle, the development of these alloys would have to entirely depend on empiricism.

In the above process, we have also delineated the phase relations as a function of temperature and cation; such knowledge has allowed us to understand the phenomena of "reverse transformation" which has been extensively discussed in the recent literature. We have shown that such reverse transformation can be easily understood by the temperature dependence of the phase boundaries. Such dependence is especially strong for larger cations. The cations investigated in our research include rare-earth ions of various sizes and alkali earth ions. These cations can be used singly or in combination.

In the area of melilite-containing silicon nitride, we have established the phase relations between melilite and various neighboring phases in the silicon nitride rich corner. We have determined the solubility limit and the stability of melilite as a function of rare-earth cation additives. We have used melilite as an effective second phase to aid sintering of silicon nitride and we have found satisfactory mechanical properties of this series of composites. In the area of superplastic silicon nitride, we have investigated a series of mixed a/b silicon nitride with fine grains and achieved superplastic forming at relatively low temperatures and high rates. The relation between flow stress and fracture stress has been used to correlate the formability of these materials. Finally, all the superplastic silicon nitrides exhibit higher room temperature bend strength after post-forming annealing. Thus, transient superplastic deformation does not impair the ultimate mechanical properties of the materials.

V. PERSONNEL SUPPORTED

This grant supports US graduate students (Anatoly Rosenflanz and Jack Smith) toward thesis research.

VI. PUBLICATIONS

(a) A. Rosenflanz and I-Wei Chen, "Phase Relationships and Stability of α'-SiAlON," accepted for publication in J. Amer. Ceram. Soc. (1998)

(b) A. Rosenflanz and I-Wei Chen, "Kinetics of Phase Transformations in SiAlON Ceramics I. Effects of Cation Size, Composition and Temperature," submitted to J. Europ. Ceram. Soc. (1998)

(c) A. Rosenflanz and I-Wei Che, "Kinetics of Phase Transformations in SiAlON Ceramics II. Reaction Paths," submitted to J. Europ. Ceram. Soc. (1998)

(c) Z.-K. Huang, A. Rosenflanz, and I-W. Chen, "SiAlON Composites Containing Rare-Earth Melilite and Neighboring Phases," *J. Amer. Ceram. Soc.*, **79** [8] 2081-90 (1996).

(d) Z.-K. Huang, A. Rosenflanz, and I-W. Chen, "Pressureless Sintering of Si_3N_4 Ceramics Using AlN and Rare Earth Oxides," *J. Amer. Ceram. Soc.*, **80** [5] 1256-62 (1997).

(e) A. Rosenflanz and I-W. Chen, "Classical Superplasticity in SiAlON Ceramics," *J. Amer. Ceram. Soc.*, **80** [6] 1341-52 (1997).

(f) I-Wei Chen and A. Rosenflanz,"A Tough SiAlON Ceramic Based on alpha Si_3N_4 with a Whisker-like Microstructure" Nature, 389, 701-04 (1997).

VII. INVITED TALKS BY PI

"Fracture Mechanics of Fatigue in Ceramics," at the 98th Annual Meeting of The American Ceramic Society, Indianapolis, IN, April, 1996.

"Ceramic Superplasticity," at the Workshop on High-Temperature Deformation of Structural Materials, Schloss Ringberg, Tegernsee, Germany, July, 1996.

"Phase Transformations of Silicon Nitride and Silicon Carbide," at the Workshop on Grain Boundary Dynamics of Precursor-Derived Covalent Ceramics, Schloss Ringberg, Tegernsee, Germany, November, 1996.

"Ceramic Superplasticity," Workshop on Atomistic Simulation of Crystal Plasticity and Fracture, Argonne, IL, August 1997.

"Innovations in Processing of Structural Ceramics with High Aspect Ratio Features," International Symposium on Novel Synthesis and Processing of Ceramics, Kurume, Japan, October 1997.

"Paradigms of Innovations in Materials Research," plenary lecture, Annual Meeting of the Chinese Society for Materials Science, Tainan, Taiwan, Novemeber 1997.

"Phase Relations and Phase Stability in SiAlON Systems," Annual Meeting of the American Ceramic Society, Cincinnati, OH, May 1998.

"Issues and Possible Solutions for Silicon Nitride used below 1400^{o}C," Workshop on Ultrahigh Temperature Ceramics, Boulder, CO, May 1998.

"In-Situ Toughened Alpha SiAlON," Conference on the New Developments in High Temperature Ceramics, Istanbul, Turkey, August 1998.

"In-Situ Toughened Alpha SiAlON," First International Conference on Inorganic Materials, Versailles, France, September 1998.

VIII. TRANSITIONS, PATENTS, AND HONORS

Samples of in-situ toughened α'-SiAlON have been provided to a leading US cutting tool manufacturer and a leading German cutting tool manufacturer to evaluate their cutting performance. This evaluation process is on-going as the cutting applications are material specific, depending on the work piece as well as the tool. Thus, composition, microstructure and property optimization is required for each cutting application.

Several patent applications on in-situ toughened α'-SiAlON have been filed at the US Patent Office. International patents have also been filed.

Professor I-Wei Chen received Alexander von Humboldt Prize that is awarded to senior US scientists by the Alexander von Humboldt Foundation of Germany.

Mr. Anatoly Rosenflanz received PhD from the University of Michigan and is now a staff researcher at 3M Research Center at Minneapolis, MN.

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13. ABSTRACT *(Maximum 200 words)*
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